The Carriers of the Interstellar Unidentified Infrared Emission Features: Aromatic or Aliphatic?

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ABSTRACT

The unidentified infrared emission (UIE) features at 3.3, 6.2, 7.7, 8.6, and $11.3\,\mu$m, commonly attributed to polycyclic aromatic hydrocarbon (PAH) molecules, have been recently ascribed to coal- or kerogen-like organic nanoparticles with a mixed aromatic-aliphatic structure. However, we show in this *Letter* that this hypothesis is inconsistent with observations. We estimate the aliphatic fraction of the UIE carriers based on the observed intensities of the $3.4\,\mu$m and $6.85\,\mu$m emission features by attributing them exclusively to aliphatic C–H stretch and aliphatic C–H deformation vibrational modes, respectively. We derive the fraction of carbon atoms in aliphatic form to be $< 15\%$. We conclude that the UIE emitters are predominantly aromatic, with aliphatic material at most a minor part of the UIE carriers. The PAH model is consistent with astronomical observations and PAHs dominate the strong UIE bands.

Subject headings: dust, extinction — ISM: lines and bands — ISM: molecules

1.   Introduction

The "unidentified infrared emission" (UIE) bands, a distinct set of spectral features at wavelengths of 3.3, 6.2, 7.7, 8.6, 11.3 and $12.7\,\mu$m, dominate the mid-infrared spectra of many bright astronomical objects. They are ubiquitously seen in the interstellar medium (ISM) of our own galaxy and star-forming galaxies, both near and far, and account for over 10% of their total infrared (IR) luminosity (see Joblin & Tielens 2011). Although the exact nature of the carriers remains unknown, the UIE bands are commonly attributed to polycyclic aromatic hydrocarbon (PAH) molecules (Léger & Puget 1984, Allamandola et al. 1985). The identification of the UIE bands is important as they are a useful probe of the cosmic star-formation history, and their carriers are an essential player in galactic evolution.

Very recently, Kwok & Zhang (2011; hereafter KZ11) argue that the UIE bands arise from coal- or kerogen-like organic nanoparticles, consisting of chain-like aliphatic hydrocarbon material

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linking small units of aromatic rings. This hypothesis has potentially important implications for our understanding of stellar evolution, interstellar chemistry, and the formation of our solar system. If confirmed, it would establish an important link among stars at their late evolutionary stages, the ISM, and the solar system, as the kerogen-like organic matter seen in meteorites (Derenne & Robert 2010; Cody et al. 2011) has similar chemical structures as those suggested by KZ11 for the UIE bands seen in the ISM and in circumstellar environments around evolved stars (i.e., planetary nebulae and proto-planetary nebulae).

However, the KZ11 hypothesis of substantially aliphatic organic matter as the UIE carriers does not appear to be consistent with the observed strengths of the UIE bands. As will be elaborated below in §2 and §3, astronomical observations show that if aliphatic hydrocarbon units are present in the UIE carriers, they must be a minor constituent. Further, their arguments against the PAH model do not seem to pose a problem (see §4).

2. Constraints on the Aliphatic Fraction from the $3.4\,\mu$m Feature

KZ11 argue that the material responsible for the UIE features has a substantial aliphatic component, based on the mid-IR spectra of NGC 7027 (a planetary nebula), IRAS 22272+5435 (a protoplanetary nebula), and the Orion bar (a photodissociation region in the Orion nebula). They decompose the 3–$20\,\mu$m spectra of these objects into three components: the UIE bands, broad plateaus (several μm in width) peaking at 8 and $12\,\mu$m, and a thermal continuum. They attribute the broad plateau features (which account for $\sim 1/3$ of the 3–$20\,\mu$m power of these objects) to aliphatic branches of the UIE carriers, similar to the coal model for the UIE bands (Guillois et al. 1996). Recognizing the challenge of the coal model in being heated to emit the UIE bands (Puget et al. 1995), KZ11 hypothesize that the coal-like UIE carriers are nanometer in size or they are heated by the chemical energy released from the $H + H \rightarrow H_2$ reaction (Duley & Williams 2011).

Aliphatic hydrocarbon has a band at $3.4\,\mu$m due to the C–H stretching mode (Pendleton & Allamandola 2002). In some HII regions, reflection nebulae and planetary nebulae (as well as extragalactic regions, e.g., see Yamagishi et al. 2012, Kondo et al. 2012), the UIE near $3\,\mu$m exhibits a rich spectrum: the dominant $3.3\,\mu$m feature is usually accompanied by a weaker feature at $3.4\,\mu$m along with an underlying plateau extending out to $\sim 3.6\,\mu$m. In some objects, a series of weaker features at 3.46, 3.51, and $3.56\,\mu$m are also seen superimposed on the plateau, showing a tendency to decrease in strength with increasing wavelength (see Figure 1 and Geballe et al. 1985, Jourdain de Muizon et al. 1986, Joblin et al. 1996). While assignment of the $3.3\,\mu$m emission feature to the aromatic C–H stretch is widely accepted, the precise identification of the $3.4\,\mu$m feature (and the accompanying weak features at 3.46, 3.51, and $3.56\,\mu$m and the broad plateau) remains somewhat controversial. By assigning the $3.4\,\mu$m emission exclusively to aliphatic C–H, one can place an upper limit on the aliphatic fraction of the emitters of the UIE features.

Let $I_{3.4}$ and $I_{3.3}$ respectively be the observed intensities of the $3.4\,\mu$m and $3.3\,\mu$m emission

features. In interstellar and circumstellar environments, $I_{3.4}/I_{3.3}$ typically ranges from ~ 0.06 to ~ 0.20, depending on the local conditions (Schutte et al. 1993). Let $A_{3.4}$ and $A_{3.3}$ respectively be the band strengths of the aliphatic and aromatic C–H bonds. We take $A_{3.4} = 2.7 \times 10^{-18}$ cm per aliphatic C–H bond, averaged over ethane, hexane, ethyl-benzene, and methyl-cyclo-hexane (d'Hendecourt & Allamandola 1986, Muñoz-Caro et al. 2001).[1] We take $A_{3.3} = 4.0 \times 10^{-18}$ cm per aromatic C–H bond for small neutral PAHs (Draine & Li 2007).

Let $N_{\rm H,aliph}$ and $N_{\rm H,arom}$ respectively be the numbers of aliphatic and aromatic C–H bonds in the emitters of the $3.3\,\mu$m UIE feature. We obtain $N_{\rm H,aliph}/N_{\rm H,arom} \approx (I_{3.4}/I_{3.3}) \times (A_{3.3}/A_{3.4}) \approx 0.30$, taking $I_{3.4}/I_{3.3} = 0.2$ [KZ11 estimate $I_{3.4}/I_{3.3} \approx 0.22$ for NGC 7027, and $I_{3.4}/I_{3.3} \approx 0.19$ for the Orion bar]. We assume that one aliphatic C atom corresponds to 2.5 aliphatic C–H bonds (intermediate between methylene -CH_2 and methyl -CH_3) and one aromatic C atom corresponds to 0.75 aromatic C–H bond (intermediate between benzene C_6H_6 and coronene $C_{24}H_{12}$). Therefore, in the UIE carriers the ratio of the number of C atoms in aliphatic units to that in aromatic rings is $N_{\rm C,aliph}/N_{\rm C,arom} \approx 0.30 \times (0.75/2.5) = 0.09$, showing that the aliphatic component is only a minor part of the UIE emitters. KZ11 take $I_{3.4}/I_{3.3} \approx 1.88$ for the protoplanetary nebula IRAS 22272+5435 (but much smaller $I_{3.4}/I_{3.3}$ ratios have also been reported for this source; see Goto et al. 2003). So far only a few sources (exclusively protoplanetary nebulae) are reported to have $I_{3.4}/I_{3.3} \gtrsim 1$ (Hrivnak et al. 2007). They are atypical UIE sources: their UIE spectra have most of the power emitted from two broad bands peaking at $\sim 8\,\mu$m and $\sim 11.8\,\mu$m, while typical UIE spectra have distinctive peaks at 7.7, 8.6, and $11.3\,\mu$m (see Tokunaga 1997).[2]

We note that $N_{\rm C,aliph}/N_{\rm C,arom} = 0.09$ is an upper bound as the $3.4\,\mu$m emission feature could also be due to anharmonicity of the aromatic C–H stretching mode (Barker et al. 1987). Let ν be the vibrational quantum number. In a harmonic oscillator, the level spacing is constant; the $\Delta\nu = 1$ transition between high ν levels results in the same spectral line as for the $\nu = 1 \rightarrow 0$ transition. Anharmonicity decreases the spacing between the higher ν levels, and the $\Delta\nu = 1$ transitions between higher ν levels occur at longer wavelengths. The anharmonicity model explains the weaker features (at $3.40\,\mu$m, $3.51\,\mu$m, ...) as "hot bands" ($\nu = 2 \rightarrow 1$, $\nu = 3 \rightarrow 2$, ...) of the $3.3\,\mu$m fundamental $\nu = 1 \rightarrow 0$ aromatic C–H stretching mode. The $3.4\,\mu$m emission feature could also be due in part to "superhydrogenated" PAHs in which some peripheral C atoms have two H atoms

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[1]Typical type II Kerogens have $A_{3.4} \approx 2.8 \times 10^{-18}$ cm per C atom while the $3.3\,\mu$m aromatic feature is barely visible (see Figure 2 in Papoular 2001). This clearly shows that kerogen – at least this type – is not a good explanation for the $3.3\,\mu$m and $3.4\,\mu$m emission features. In coals, the $3.3\,\mu$m aromatic feature is usually weaker than the $3.4\,\mu$m aliphatic feature except for those with high ranks (i.e., more evolved, more ordered, with lower H/C and O/C ratios). As coal evolves, the progressive release of heteroatoms (decreasing H/C and O/C) leads to formation of planar clusters of benzene-type rings followed by stacking of these aromatic sheets to form disordered stacks of graphitic planes (see Papoular 2001). As a result of the progressive aromatization, $A_{3.4}/A_{3.3}$ decreases as coal evolves. The aliphatic C–H deformation band at $6.85\,\mu$m band disappears in highly evolved coal (e.g., anthracite, see Papoular 2001).

[2]Overall, the IR spectra of coals or kerogens resemble that of atypical sources (e.g., some protoplanetary nebulae; see Guillois et al. 1996). They do not resemble the UIE features seen in the interstellar sources except for highly evolved (i.e., highly aromatized) coals (see Papoular 2001).

(see Figure 2). The extra H atom converts the originally aromatic ring into an aliphatic ring. This creates two aliphatic C–H stretching bands: one due to the symmetric and the other to the asymmetric C–H stretching modes. These bands would fall near $3.4\,\mu$m and $3.5\,\mu$m, with the former more intense than the latter, consistent with astronomical observations (Bernstein et al. 1996). The $3.4\,\mu$m feature may also result from aliphatic sidegroups attached as functional groups to PAHs (see Figure 2; Duley & Williams 1981, Pauzat et al. 1999, Wagner et al. 2000). The C–H stretching modes of methyl (-CH$_3$), methylene (-CH$_2$-), and ethyl (-CH$_2$CH$_3$) sidegroups on PAHs fall near the weaker satellite features associated with the $3.3\,\mu$m band. All these possibilities (i.e., anharmonicity, superhydrogenation, and aliphatic sidegroups) probably contribute to the $3.4\,\mu$m emission, the extent of each depending on conditions in the local environment. Sandford (1991) argued that the satellite features at 3.40, 3.46, 3.51, and $3.56\,\mu$m in NGC 7027 cannot be predominantly due to aliphatic sidegroups on PAHs.

KZ11 note that the $3.4\,\mu$m aliphatic C–H stretching mode is commonly observed in *absorption* in the diffuse ISM. If the UIE carriers have the same mixed aromatic-aliphatic structure as the bulk of the hydrocarbon material, then in heavily obscured regions, both the $3.3\,\mu$m band and the $3.4\,\mu$m band would show up in *absorption*, with the $3.4\,\mu$m absorption band much *weaker* than the $3.3\,\mu$m absorption band. However, astronomical observations have actually shown the opposite (see Figure 1): the $3.4\,\mu$m absorption band is much *stronger* than the $3.3\,\mu$m absorption band (e.g. in the Galactic center source GCS 3, the $3.4\,\mu$m absorption band is stronger than the $3.3\,\mu$m band by a factor of 35; Chiar et al. 2000). Therefore, the bulk of the $3.4\,\mu$m absorber in the ISM must be hydrocarbon material in the larger grains, evidently more strongly aliphatic than the UIE carriers (Dartois et al. 2007).

3. Constraints from the $6.85\,\mu$m Feature

In addition to the $3.4\,\mu$m C–H stretching mode, aliphatic hydrocarbon materials also have two C–H deformation bands at $6.85\,\mu$m and $7.25\,\mu$m.[3] These two bands have been observed in weak absorption in the diffuse ISM (Chiar et al. 2000). They are also seen in emission in interstellar and circumstellar UIE sources. Their strengths (relative to the nearby $7.7\,\mu$m C–C stretching band) also allow an estimate of the aliphatic fraction of the UIE carrier.[4]

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[3]One may argue that in the KZ11-type coal- or kerogen-like material, the aliphatic C–H bands may not occur at the same wavelengths as for pure aliphatics or PAHs with simple aliphatic sidegroups: the aliphatic H atoms occupy a broad range of local chemical environments, subject to hydrogen bonding perturbations by nearby O and S atoms. Such interactions could conceivably shift the C–H frequencies from their "normal" aliphatic positions. However, laboratory measurements have shown that the aliphatic C–H bands in coal or kerogen do occur at $3.4\,\mu$m and $6.85\,\mu$m, displaying little wavelength shift compared to that of pure aliphatics (see Papoular 2001).

[4]Coals or kerogens do not exhibit a distinct band at $7.7\,\mu$m and thus one cannot infer their aliphatic fractions from $I_{6.85}/I_{7.7}$.
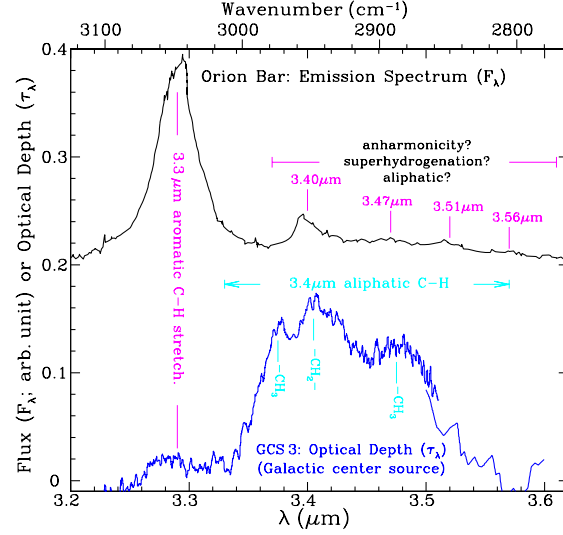
Fig. 1.— Comparison of the 3.15–3.65 μm *emission* spectrum of the Orion Bar (position 4; black; Sloan et al. 1997) with the optical depth (*absorption*) spectra of GCS 3 (a Galactic center source; blue; Chiar et al. 2000). The weakness of the 3.4 μm feature in the *emission* spectrum indicates that the *aliphatic* component must be minor, even assuming that the 3.4 μm emission is exclusively due to aliphatic C–H (i.e., neglecting anharmonicity and superhydrogenation). In contrast, the *absorption* spectrum of the diffuse ISM toward GCS 3 is dominated by aliphatic hydrocarbon.
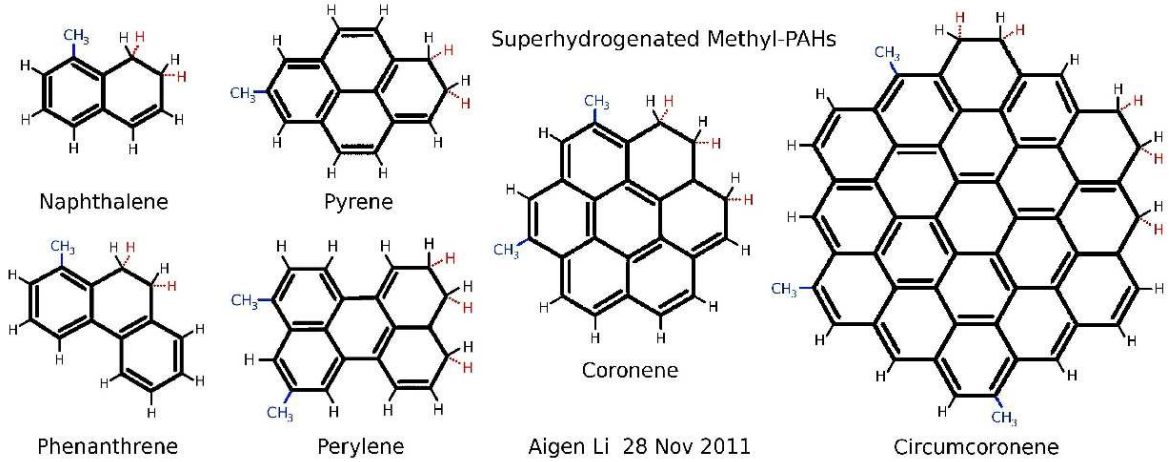

Fig. 2.— Examples of "superhydrogenated" PAHs with methyl (-CH$_3$) aliphatic sidegroups. In addition to anharmonicity, superhydrogenation and methyl-like aliphatic sidegroups attached to PAHs may contribute to the weak 3.4 μm emission feature accompanying the 3.3 μm feature.

Let $I_{6.85}$ and $I_{7.7}$ be the observed intensities of the $6.85\,\mu$m and $7.7\,\mu$m emission features. Let $A_{6.85}$ and $A_{7.7}$ be the strengths of the $6.85\,\mu$m aliphatic C–H band and the $7.7\,\mu$m aromatic C–C band. We take $A_{6.85} = 2.3 \times 10^{-18}$ cm per CH_2 or CH_3 group, an average of that measured for methylcylcohexane ($A_{6.85} = 3.0 \times 10^{-18}$ cm per CH_3 group; d'Hendecourt & Allamandola 1986) and for hydrogenated amorphous carbon ($A_{6.85} = 1.5 \times 10^{-18}$ cm per CH_2 or CH_3 functional group; Dartois & Muñoz-Caro 2007).[5]

We take $A_{7.7} = 5.4 \times 10^{-18}$ cm per C atom for charged aromatic molecules (Draine & Li 2007). Let $N'_{\rm C,aliph}$ and $N'_{\rm C,arom}$ respectively be the numbers of aliphatic and aromatic C atoms in the emitters of the 6–8 μm UIE bands. Let $B_\lambda(T) \propto \lambda^{-3}/\left[\exp\left(hc/\lambda kT\right) - 1\right]$ be the Planck function at wavelength λ and temperature T (where h is Planck's constant, c is the speed of light, and k is Boltzmann's constant), with $B_{6.85}/B_{7.7} \approx 0.9 \pm 0.2$ for $330 < T < 1000\,$K. Then $N'_{\rm C,aliph}/N'_{\rm C,arom} \approx (I_{6.85}/I_{7.7}) \times (A_{7.7}/A_{6.85}) \times (B_{7.7}/B_{6.85}) \approx 0.10$ for NGC 7027 ($I_{6.85}/I_{7.7} \approx 0.039$) and $N'_{\rm C,aliph}/N'_{\rm C,arom} \approx 0.14$ for the Orion bar ($I_{6.85}/I_{7.7} \approx 0.053$). KZ11 derived $I_{6.85}/I_{7.7} \approx 1.43$ for IRAS 22272+5435, but the observed spectrum and decomposition fit support a significantly smaller value.

We conclude that the carriers of the 6–8 μm UIE bands are predominantly aromatic, with $< 15\%$ of the C atoms in aliphatic form. The aliphatic fraction, while still small, appears to be higher than estimated for the 3.3–3.4 μm band carriers, consistent with increased aromatization of the smallest particles, which are heated to the highest temperatures.

4. Discussion

KZ11 attribute the broad plateau emission around 8 and 12 μm to the aliphatic component of the UIE carreirs. They hypothesize that the clustering of aromatic rings may break up the simple methyl- or methylene-like sidegroups and hence the aliphatic components may take many other forms (e.g., -CH=CH_2, -CH=CH-, C=CH_2, C=C-H). They speculate that the in-plane and out-of-plane bending modes of these sidegroups may combine to form the plateau. We note that the PAH model naturally accounts for the so-called "plateau" emission through the combined wings of the C–C and C–H bands. We also note that the clustering of aromatic rings and aliphatic chains would be accompanied by forming new C–C bonds and losing H atoms. Laboratory measurements of coals have shown that lowering the H content leads to aromatization (see Papoular 2001).

KZ11 claim that the PAH hypothesis postulates that the UIE emission is excited exclusively by far-UV photons, and that this is inconsistent with observation of UIE emission in reflection nebulae excited by cool stars (Sellgren et al. 1990). However, Li & Draine (2002) explicitly considered the

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[5]Typical type II Kerogens have $A_{6.85} \approx 3.6 \times 10^{-19}$ cm per C atom (Papoular 2001). If 15% of the C atoms in kerogens are in aliphatic form, for kerogens we would have $A_{6.85} \approx 2.4 \times 10^{-18}$ cm per aliphatic C atom. This is close to that adopted in this work: $A_{6.85} \approx 2.3 \times 10^{-18}$ cm per aliphatic C atom.

excitation of PAHs by longer-wavelength photons, and showed that the light from even relatively cool stars can excite UIE emission, consistent with observations. The excitation of PAHs by visible or even near-IR photons with wavelengths up to ~ 1–$2\,\mu$m has been further experimentally verified (Mattioda et al. 2005). KZ11 note the constancy of UIE band ratios in regions (e.g. the Carina nebula) where the radiation intensity changes by orders of magnitude. This is *precisely* what one expects if the emission comes from single-photon heating of nanoparticles [see Figure 13 of Li & Draine (2001), Figure 4b of Li & Draine (2002), Figure 1f of Draine & Li (2007)].

KZ11 note that of the more than 160 molecules identified in circumstellar and interstellar environments, none is a PAH. This is true, but also not surprising because the mid-IR UIE bands – the major observational information – are representative of functional groups and do not fingerprint individual PAH molecules.[6] KZ11 argue that the carrier of the UIR features cannot be a "pure aromatic compound". Proponents of the identification of the astronomical UIE features as coming from PAHs do not claim that the emitting material is "pure aromatic compound", as strictly defined by a chemist. The astronomical material may well include a *minor* aliphatic component, as well as defects, substituents (e.g., N in place of C), partial dehydrogenation, and sometimes superhydrogenation (Tielens 2008). Some of the nanoparticles may be multilayer aggregates of PAHs.

KZ11 state that PAH molecules have strong and narrow absorption features in the UV whereas the search for characteristic absorption features of PAHs superposed on the interstellar extinction curves was not successful (e.g., see Clayton et al. 2003). For individual *small* PAHs, this is true. However, in the PAH hypothesis it is natural to expect that there will be a large number of distinct species present in the ISM, and no single UV band may be strong enough to be identified in the UV. This also explains why laboratory-measured spectra of *individual* PAHs do not precisely match the observed UIE features in band widths and peak wavelengths, while *combined* laboratory spectra of neutral PAHs and their ions can successfully reproduce the UIE bands associated with many different interstellar objects (Allamandola et al. 1999). There are, in fact, over 400 diffuse interstellar bands (DIBs) in the optical that remain to be identified (Sarre 2006, Salama et al. 2011). Many of these may eventually be found to be produced by specific PAHs, but at this time we lack the laboratory spectroscopy to make the identifications. The lack of identification of any specific PAH is not a fatal problem for the PAH hypothesis, at least at this time. As we develop a better knowledge of the gas-phase spectroscopy of the larger PAHs, this story may change. If the DIBs are electronic transitions of PAHs, they hold great promise for identifying specific PAH molecules, as the electronic transitions are more characteristic of a specific PAH molecule than the

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[6]The far-IR bands are more sensitive to the skeletal characteristics of a molecule, and hence are more diagnostic of the molecular identity and more powerful for chemical identification of unknown species. In principle, far-IR spectroscopy could be used to test the KZ11 hypothesis: the KZ11-type material has an extremely "floppy" structure compared to the more rigid PAHs, and therefore there would be many low frequency skeletal bends and very low-frequency pseudo-rotations about bond axes. However, there is little information on the far-IR spectroscopy of coal or kerogen. Even for PAHs, this information is very limited (e.g., see Joblin et al. 2009, Zhang et al. 2010).

mid-IR C–H and C–C vibrational bands.

The strong interstellar 217.5 nm extinction bump is likely to be a *blend* of $\pi-\pi^*$ absorption bands from the entire population of PAHs, with the fine structures from individual PAH molecules smoothed out. Furthermore, internal conversion may lead to extreme broadening of the UV absorption bands in larger PAHs, which may account for absence of recognizable absorption features shortward of 200 nm. This has been demonstrated both experimentally and theoretically. Léger et al. (1989) measured the absorption spectra of mixtures of over 300 neutral PAH species with ~ 12–28 C atoms. Joblin et al. (1992) measured the absorption spectra of neutral PAH mixtures containing ~ 14–44 C atoms. All these spectra have a strong UV feature around 217.5 nm (but relatively broader than the interstellar bump). While it is true that laboratory PAH samples do not precisely reproduce the observed profile of the 217.5 nm feature (Léger et al. 1989, Joblin et al. 1992), this is probably due to the fact that laboratory studies are generally limited to small PAHs while interstellar PAHs are much larger (e.g. models that reproduce the ~ 3–20 μm UIE bands have most of the PAH mass in PAHs with >100 C atoms, see Li & Draine 2001, Draine & Li 2007). Indeed, Steglich et al. (2010) showed that larger PAHs indeed provide better fits to the observed 217.5 nm feature. Cecchi-Pestellini et al. (2008) also showed that a weighted sum of 50 neutral and ionized PAHs in the size range of ~ 10–66 C atoms can reproduce the 217.5 nm extinction bump observed in various environments.

5. Conclusion

We examine the hypothesis of mixed aromatic-aliphatic organic matter as the UIE carriers. We place an upper limit on the aliphatic fraction of the UIE carriers based on the observed weak intensities of the 3.4 μm and 6.85 μm emission features. By attributing them *exclusively* to aliphatic C–H stretch and aliphatic C–H deformation, we derive the fraction of carbon atoms in aliphatic form to be $< 15\%$. We conclude that the UIE emitters are predominantly aromatic: PAHs dominate the principal UIE bands. Our expectation is that confirmation will not come until we have laboratory spectroscopy of PAH candidates in the gas phase that precisely match some of the observed DIBs.

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